

Supplementary Information for

In-situ growth of Graphdiyne on Arbitrary Substrates with a Controlled-release Method

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Materials

Unless otherwise stated, the reagents and solvents used in this work were commercially obtained and used without further purification.

Substrates pretreatment

Si and SiO₂ wafers and glass slides were cut into slices about 1 cm², and then pretreated by sonicating in 3M HCl and acetone and ethanol, respectively, for 15 min, dried under a flow of argon and plasma processed to get a fresh surface and used immediately for growing graphdiyne films.

Preparation of PVP/Cu(OAc)₂ solution and PVP/Cu(OAc)₂ film

0.3 g of PVP and 0.1, 0.2, 0.3, 0.4 g of cupric acetate was dissolved in 20 mL deionized water, respectively, to get a series of slow-release solution with various Cu²⁺ concentration. 15 μL of as prepared PVP/Cu(OAc)₂ solution was dropped on the prepared sheet substrates, followed by spinning at 3000 rpm to fabricate a PVP/Cu(OAc)₂ film. For the powder substrates, the substrates were immersed in PVP/Cu(OAc)₂ solution for 3 hours, then filtered and dried to form a PVP/Cu(OAc)₂ film.

Preparation of HEB precursor solution

HEB was synthesized according to the reported synthetic route.¹

Preparation of GDY on arbitrary substrate

The substrates covered with PVP/Cu(OAc)₂ film was immersed into HEB solution with acetone and pyridine (v: v = 20: 1) as solvent, followed by keeping 3 days' standing in atmosphere environment at room temperature. Finally, bulk GDY was uniformly fabricated on the target substrates.

Characterization of Materials

The morphologies of the materials were characterized by field emission SEM (HITACHI S-4800) equipped with an energy dispersive X-ray spectrometry. TEM and HRTEM images were obtained using TEM (JEOL 2011F and 2100F). FTIR was recorded on a Bruker VECTOR 22 spectrometer in the frequency range of 4000–500 cm⁻¹. Raman spectra were recorded at the room temperature using a Thermo Scientific DXRxi system with a 532 nm laser. XPS analysis was performed by using VG Scientific ESCALab220i-XL X-ray photoelectron spectrometer with K α radiation as the excitation sources. XRD patterns were

recorded on X-ray powder diffractometer (D8 ADVANCE) with Cu K α radiation. The typical current–voltage (I–V) curves of as-prepared GDY film was recorded using a Keithley 4200 semiconductor characterization system and Signatone probe station.

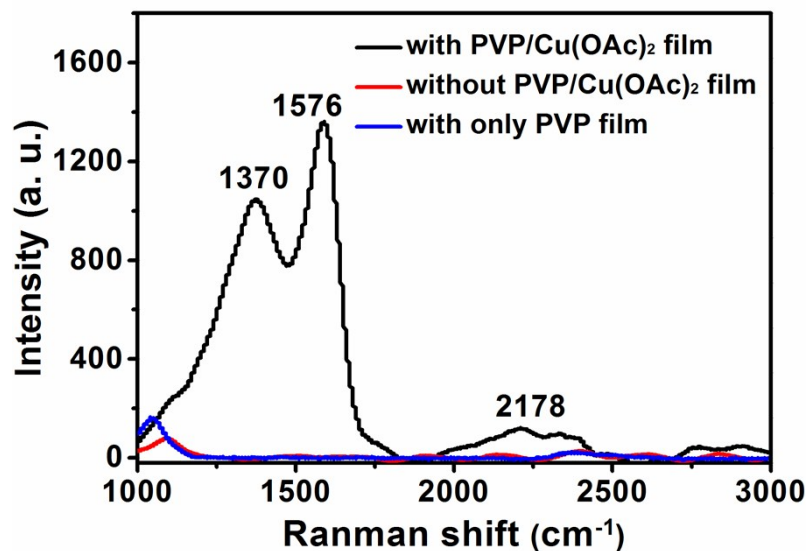


Figure S1 Typical Raman spectra of glasses growing GDY with and without PVP/ copper(II) acetate film.

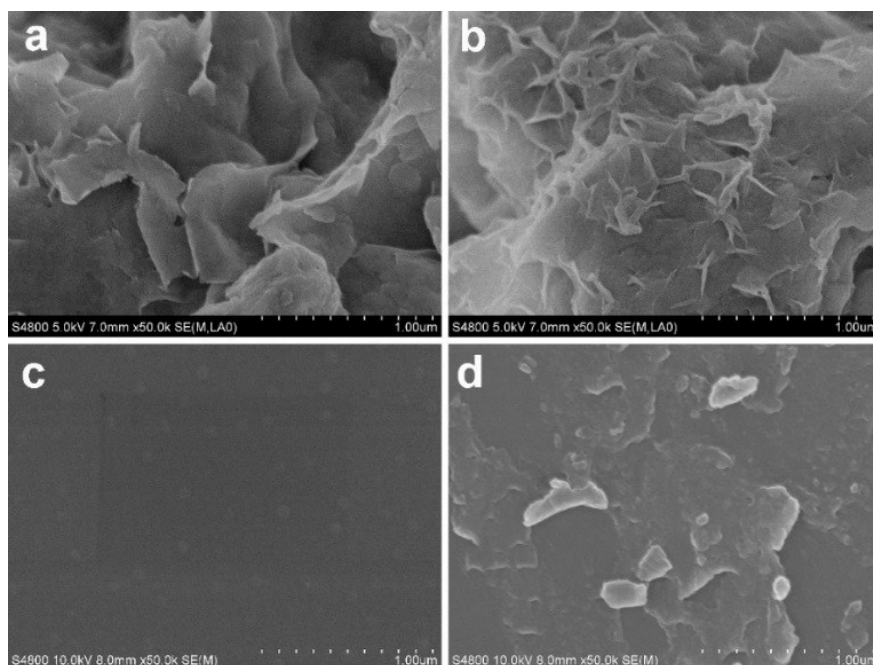


Figure S2 SEM images of different substrates before and after the fabrication of GDY: a, b) on nano-layered montmorillonite, c, d) on ITO glass.

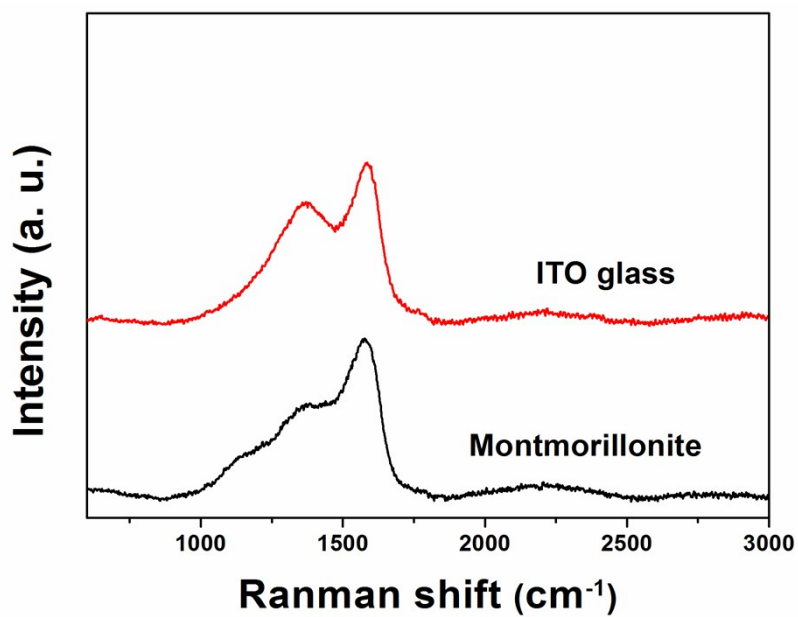


Figure S3 Typical Raman spectra of GDY fabricated on ITO glass and montmorillonite substrates.

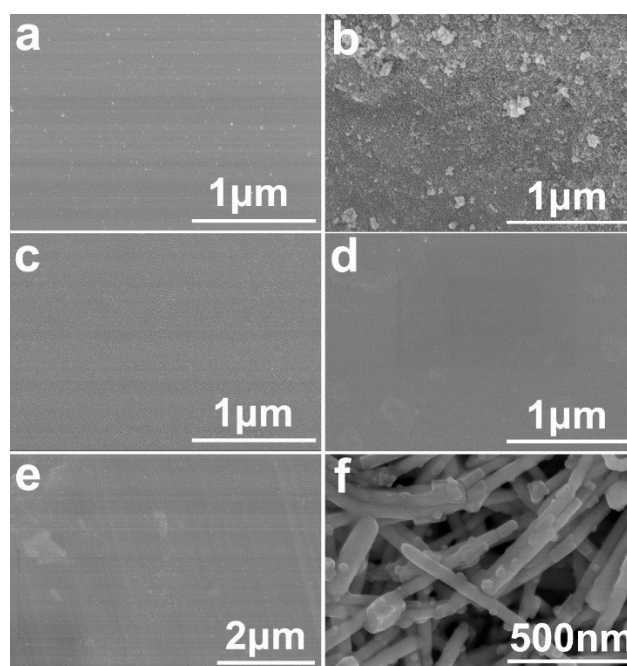


Figure S4 SEM images of different substrates. a) glass sheets, b) macro-porous silica gel, c) silicon wafers, d) ZnO film, e) Al foil and f) Ag nanowires.

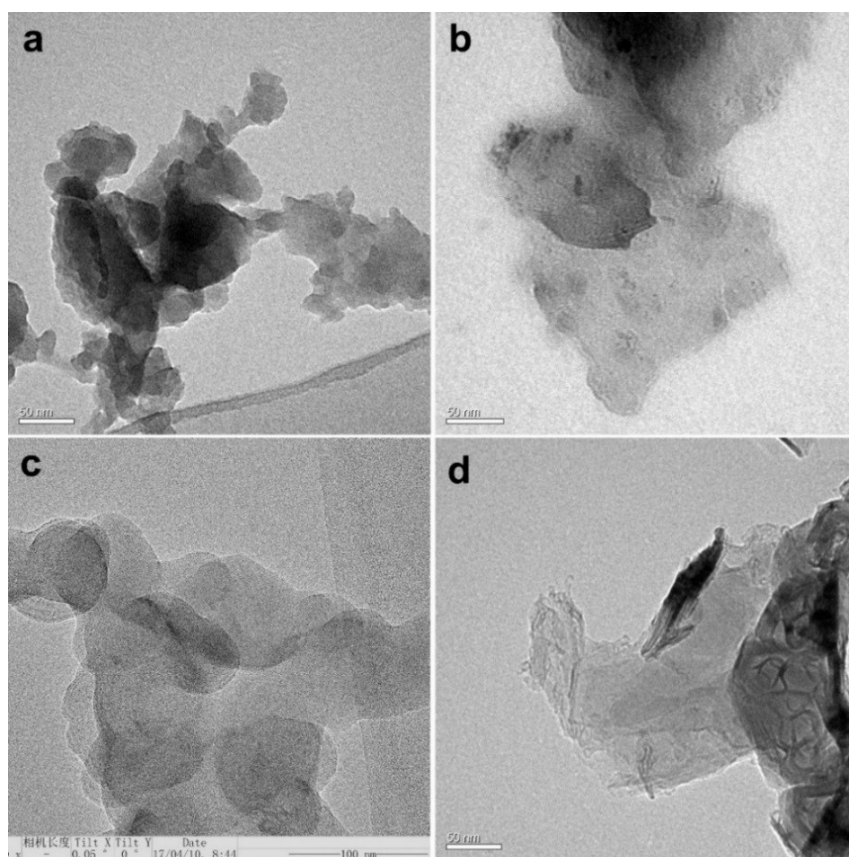


Figure S5 TEM images of as-prepared GDY peeled from (a) glass sheet, (b) macro-porous silica gel, (c) silicon wafer and (d) ZnO film.

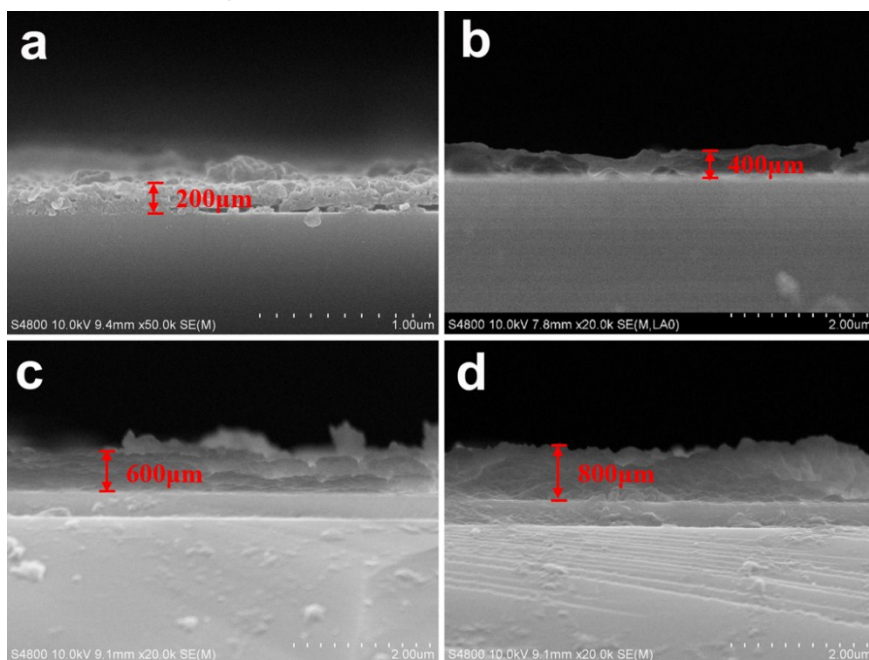


Figure S6 Thickness measurement of GDY fabricated on SiO₂ wafer by SEM.

Supplementary References

1. G. Li, Y. Li, H. Liu, Y. Guo, Y. Li and D. Zhu, *Chem. Commun.*, 2010, **46**, 3256-3258.